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Magnetic Eigenmodes in QD-Based Resonant Active Composites

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Abstract

The phenomenon of light confinement in an isolated quantum dot, provided by the resonant nature of exciton in QD and diffraction of electromagnetic waves at the dot boundary, is discussed. It has been shown that at a certain condition the quantum dot behaves as a microcavity those eigenmodes manifest themselves as additional, geometrical, resonances in the quantum dot electromagnetic response. The effect of induced magnetization of quantum dot is predicted and illustrated by the example of magnetic resonances in spherical quantum dots.

1. Introduction

A fundamental breakthrough in semiconductor device physics is connected with the recent progress in the synthesis of sheets of nano-scale 3D confined narrow-gap insertions in a host semiconductor, quantum dots (QDs). In particular, it was predicted that lasers based on QDs will show radically changed characteristics as compared to conventional quantum well lasers [1, 2]. The large body of recent results on physical properties of QDs and their utilization for the QD laser design has been accumulated in a monograph [3].

The key peculiarity of QDs emerges from the 3D confinement of the charge carriers determined by QD size and shape. However, there exists a class of effects governed by the QD size and shape, which have not received much attention so far. These effects are related to resonant nature of the exciton which provides a dramatic resonant discontinuity of the dielectric function at the QD boundary and, consequently, gives rise inhomogeneity of the electromagnetic field both inside and outside QD. By analogy with charge carrier confinement, redistribution of the electromagnetic field energy between the QD interior and exterior under effect of the QD boundary can be referred to as *light confinement*. Owing to this effect, diffraction of light by QDs are expected to contribute significantly to the electromagnetic response properties of QDs. In many cases the role of diffraction can be properly accounted for the formation in QD of depolarization electromagnetic field, e.g., in dipole approximation of the diffraction theory.

To our knowledge, some physical consequences of the light confinement in an individual QD first time were considered by Schmitt-Rink *et al.* [4]. Manifestation of this phenomenon

in relation to the scanning near-field optical microscopy was discussed by Martin *et al.* [5] for geometrically complex mesoscopic systems and by Hanewinkel *et al.* [6] for QDs. An asymmetry of optical absorption and gain spectra in single QD because of depolarization field has been mentioned in Ref. [6]. Recently it has been predicted and experimentally verified that the light confinement in QD arrays constituted by anisotropically shaped QDs manifests itself as polarization splitting of the gain band [7] and, in more general case, as the fine structure of this band [8]. A concept of active composite has been introduced by Ref. [7]. A set of new effects related to the light confinement in QDs is analyzed in Ref. [9]. One of them, excitation of geometrical resonances in QD arrays, we consider here in more detail.

2. Electromagnetic Response of a Single QD

Conventional phenomenological model of the gain in a QD is based on semi-classical theory of two-level systems which gives the well-known Lorentzian polarizability of QD: $\alpha(\omega) = (g_0/\varepsilon_h)[\omega - \omega_0 + i/\tau]^{-1}$, and $\varepsilon(\omega) = \varepsilon_h[1 + \alpha(\omega)]$. Here ω_0 is the exciton resonant frequency and τ is the exciton dephasing time in QD, ε_h is the frequency-independent complex-valued permittivity of the host medium. The phenomenological parameter g_0 is proportional to the oscillator strength of the transition. Such a primitive model does not take into account effect of depolarization field which makes the polarizability tensorial for anisotropically shaped QDs and shifts exciton resonance [7], [8] (for spherical QDs $\omega_N = \omega_0 - g_0/3\varepsilon_h$). The depolarization field approximation is applicable when the condition $\kappa(\omega) = kR\sqrt{\varepsilon(\omega)} \ll 1$ holds true. Beyond the scope of this condition, when the wavelength inside QD becomes comparable with its linear extension, the role of diffraction by QD is irreducible to the effect of depolarization. Below we discuss this effect restricting ourselves to the spherical QD for simplicity.

Let an isolated spherical QD of the radius R be exposed to external electromagnetic field. The problem of wave diffraction by a sphere has been exactly solved in the early of century by using the variable separation in the spherical basis. In view of the condition $kR\sqrt{\varepsilon_h} \ll 1$, which is valid for any realistic QDs, this solution is essentially simplified [10] and presents the field outside the sphere by:

$$\begin{Bmatrix} \mathbf{E} \\ \mathbf{H} \end{Bmatrix} = (\nabla\nabla \cdot + \varepsilon_h k^2) \begin{Bmatrix} \Pi^e \\ \Pi^m \end{Bmatrix} + ik\nabla \times \begin{Bmatrix} \Pi^m \\ -\sqrt{\varepsilon_h}\Pi^e \end{Bmatrix}, \quad (1)$$

where Hertz potentials are given by:

$$\begin{Bmatrix} \Pi^e \\ \Pi^m \end{Bmatrix} = \frac{R^3}{3r} \begin{Bmatrix} \alpha^e \mathbf{E}_0 \\ \alpha^m \mathbf{H}_0 \end{Bmatrix} \exp(ik\sqrt{\varepsilon_h}r). \quad (2)$$

and the electric and magnetic polarizabilities of the sphere, $\alpha^{e,m}(\omega)$, are as follows:

$$\alpha^e(\omega) = 3 \frac{\varepsilon(\omega)F(\kappa) - \varepsilon_h}{[\varepsilon(\omega)F(\kappa) + 2\varepsilon_h](1 - ikR\sqrt{\varepsilon_h}) + i(kR\varepsilon_h)^2 F(\kappa)}, \quad (3)$$

$$\alpha^m(\omega) = 3 \frac{F(\kappa) - 1}{[F(\kappa) + 2](1 - ikR) + i(kR)^2 F(\kappa)}. \quad (4)$$

The function

$$F(\kappa) = 2 \frac{\sin \kappa - \kappa \cos \kappa}{(\kappa^2 - 1) \sin \kappa + \kappa \cos \kappa} \quad (5)$$

is responsible for the diffraction effect.

It can easily be found that the depolarization field approximation comes into play in the limit $F(\kappa) \rightarrow 1$. At $|\kappa| > 1$ the wavelength inside the QD becomes comparable with its linear

extension, and, as follows from Eqs. (3)–(5), scattered wave field is generated by irradiation of both electric and magnetic dipoles indicating thus induced magnetism of QDs. Physical mechanism of magnetization of dielectrics with linear extension compared with the internal wavelength is related to the excitation of internal TE_{1q1} cavity modes ($q = \pm 1, \pm 2, \dots$ are the polar indices of the modes) in scattering object, which thus behaves itself as a microcavity. Such modes give rise to a curl electric current in its turn inducing nonzero magnetic moment of the object [11]. The given effect is known in macroscopic electrodynamics; it is observed in macroscopic dielectric composite materials [12]. A peculiarity of the magnetism in QDs is its pronounced resonant nature. The eigenmodes indicated are called geometrical resonances. The term "geometrical" [10] is related to that the resonances occur exceptionally owing to a certain geometrical configuration of the QD.

The resonant conditions for electric and magnetic geometrical resonances are completely determined by the properties of the function $F(\kappa)$. This function demonstrates a set of resonances in the vicinity of the exciton frequency, whereas $F(\kappa) \rightarrow 1$ at $|\omega - \omega_0| \rightarrow \infty$ reducing the problem to that considered in Refs. [7], [8]. Thus, the geometrical resonances can manifest themselves in the vicinity of the exciton frequency ω_0 and certainly disappear far away this frequency region. However, concerning electrical geometrical resonances we have to conclude that they are not of interest because they can not be excited separately from the main exciton resonance ω_N . This is because both types of electric resonances are excited by electric component of the external field. Since the intensity of electrical geometrical resonance is a small portion of the main resonance intensity, its contribution results in small-amplitude beatings on the main line slope. Thus, higher electrical eigenmodes practically do not influence the main (depolarization) resonance. Unlike to electric resonances, magnetic ones are excited by magnetic component of the external field; in such situation placement of a QD in a microcavity in an antinode of magnetic field creates a possibility to make the effect evident without excitation of the main resonance.

Note also that the magnetic resonance exhibits much longer radiative lifetime as compared to the main resonance [9]. Furthermore, this lifetime is extremely longer than the intrinsic dephasing time, which therefore is crucial for possibility to observe the magnetic resonance.

3. Magnetization of QD Arrays

Occurrence of the magnetic geometrical resonance in isolated QDs must lead to magnetization of a QD array in the vicinity of the exciton frequency, essentially shifted to the blue with respect to the main resonance observable in experiments. Electromagnetic properties of composites are usually modeled in the framework of the effective-medium approach using the well-known Mossotti–Clausius formalism [12]. A homogeneous medium with effective constitutive parameters — such as conductivity, susceptibility and permittivity — is said to replace the composite. Following to conventional procedure, we present the homogenization of a QD-based composite with induced magnetic polarizabilities of inclusions.

General expression for the effective permeability tensor of a dilute composite medium comprising a regular ensemble of identical, electrically small magnetic inclusions dispersed in a host dielectric material is as follows:

$$\hat{\mu}_{eff}(\omega) = \hat{\mathbf{I}} + 4\pi f_V \hat{\alpha}^m(\omega) \left[\hat{\mathbf{I}} + f_V \hat{\delta} \hat{\alpha}^m(\omega) \right]^{-1}, \quad (6)$$

where $\hat{\delta}$ is the lattice tensor completely determined by geometry of the array, $\hat{\alpha}^m$ is the magnetic polarizability tensor of a single inclusion (for spherical QDs this tensor reduces to scalar quantity α^m (4)), f_V is the volume fraction of inclusions. The notation $\hat{\delta} \hat{\alpha}^m$ stands for the inner tensor product. Rigorous derivation of this expression based on the integral equations of macroscopic electrodynamics has been presented by Khiznjak [10]. An estimate of the array permeability can

be obtained from equation (6). Using realistic parameters, one can find $\mu - 1 \sim 0.05 - 0.1$. This is available for observation. For more correct estimate, effects of inhomogeneous broadening has to be involved in the analysis. Thus, we can conclude that the electromagnetic wave diffraction by QDs may result in manifestation by QD arrays of magnetic properties although both QD and surrounding materials are dielectric.

4. Conclusion

In this paper occurrence of magnetic geometrical resonances caused by the excitation of eigenmodes in QDs, which thus behave themselves as microcavities, is predicted. Having much smaller intensity as compared to the main exciton peak, these resonances can be evident owing to their shifts with respect to the main exciton peak and can be excited by placing of QD in a microcavity in the magnetic field antinode, where the main peak is suppressed. Measurement of the frequency shift between main exciton and magnetic resonances can be used for direct determination of the oscillator strength in QDs. In our paper we restricted ourselves to the spherical model of QD. Different QD configurations like disks or pyramids can be investigated using direct computation on the basis of the well-developed method of classical electrodynamics [13].

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